UCRL-PROC-225933

LAWRENCE LIVERMORE NATIONAL LABORATORY

Overview of Tabletop X-ray Laser Development at the Lawrence Livermore National Laboratory

J. Dunn, V. N. Shlyaptsev, J. Nilsen, R. F. Smith, R. Keenan, S. J. Moon, J. Filevich, J. Rocca, A. J. Nelson, J. R. Hunter, M. C. Marconi, Y. L. Li, A. L. Osterheld, R. Shepherd, H. Fiedorowicz, A. Bartnik, A. Ya. Faenov, T.A. Pikuz, P. Zeitoun, S. Hubert, S. Jacquemot and M. Fajardo

31 October 2006

Submitted to the Proceedings of the 10th International Conference on X-ray Lasers, Berlin, Germany 21 – 25 August, 2006

Overview of Tabletop X-ray Laser Development at the Lawrence Livermore National Laboratory

J. Dunn[1], V. N. Shlyaptsev[2], J. Nilsen[1], R. F. Smith[1], R. Keenan[1], S. J. Moon[1], J. Filevich[3], J. Rocca[3], A. J. Nelson[1], J. R. Hunter[1], M. C. Marconi[3], Y. L. Li[1*], A. L. Osterheld, R. Shepherd[1], H. Fiedorowicz[4], A. Bartnik[4], A. Ya. Faenov[5], T.A. Pikuz[5], P. Zeitoun[6], S. Hubert[7], S. Jacquemot[7] and M. Fajardo[8]

[1]Lawrence Livermore National Laboratory, Livermore, CA 94551, USA.
[2]University of California Davis-Livermore, Livermore, CA 94551, USA.
[3]NSF ERC for Extreme Ultraviolet Science and Technology and Dept. of Elect. and Comp. Engineering, Colorado State Univ., Fort Collins, USA.
[4]Inst. of Optoelectronics, Military Univ. of Technology, Warsaw, Poland.
[5]Multicharged Ions Spectra Data Center of VNIIFTRI, Moscow, Russia.
[6]Laboratoire d'Optique Appliquée, ENSTA, 91761 Palaiseau, France.
[7]Commissariat á l'Énergie Atomique, 91680 Bruyères-le-Chatel, France.
[8]Centro de Fisica dos Plasmas, Inst. Superior Técnico, Lisbon, Portugal
[*]Present address: Argonne National Laboratory, Argonne, IL 60439 USA

Summary. It is almost a decade since the first tabletop x-ray laser experiments were implemented at the Lawrence Livermore National Laboratory (LLNL). The decision to pursue the picosecond-driven schemes at LLNL was largely based around the early demonstration of the tabletop Ne-like Ti x-ray laser at the Max Born Institute (MBI) as well as the established robustness of collisional excitation schemes. These picosecond x-ray lasers have been a strong growth area for x-ray laser research. Rapid progress in source development and characterization has achieved ultrahigh peak brightness rivaling the previous activities on the larger facilities. Various picosecond soft-x-ray based applications have benefited from the increased repetition rates. We will describe the activities at LLNL in this area.

1 Introduction

By the mid-1990s there were a number of remarkable advances in soft x-ray laser experimental and theoretical research that established the viability of tabletop pumped devices. The first demonstration of a tabletop x-ray laser was reported in 1994 using the collisional excitation scheme on a fast capillary discharge apparatus [1]. The Ne-like Ar $3p - 3s$ $J = 0 - 1$ line at 46.9 nm was shown to lase and within two years was operating in saturation [2]. On the laser-driven schemes, there were a variety of approaches described. A 10 Hz Pd-like Xe ion x-ray laser scheme with $gL \sim 11$ at 41.8 nm for 40 fs irradiation of a Xe gas cell using field-induced tunneling ionization followed by collisional excitation [3]. Observation of a Ne-like titanium $3p$ - $3s$ $J = 0$ - 1 transition at 32.6 nm transient collisional excitation scheme produced high gain $g = 19$ cm^{-1} and a gain-length product of 9.5 [4]. A laser-driven recombination scheme based on the H-like Li $n = 2 - 1$ transition was also demonstrated at 13.5 nm [5]. The rapid technology advances of compact, high peak power, short pulse lasers accelerated this work and subsequent laser-pumped soft x-ray amplifiers.

This was the landscape of the tabletop x-ray lasers during 1996 when the decision at LLNL was made to start a small-scale experimental x-ray laser effort. This new approach for generating x-ray lasers with a higher repetition rate and smaller drive energy was a different direction compared to the renowned large-scale experiments using the two-beam Novette/Nova-based x-ray laser. The latter had been active for more than a decade but were now winding down as the plans for the National Ignition Facility were implemented. In this paper, we describe some of the main achievements in the tabletop x-ray laser research at LLNL over the last 10 years, as summarized in Fig.1.

2 First Experimental Steps

The transient collisional excitation scheme, as reported earlier by Shlyaptsev [6] and demonstrated by the MBI group [4], was the most appealing candidate for implementing at LLNL. It was first and foremost a collisional excitation scheme that had been well-studied by many laboratories in the Ne-like and Ni-like closed shell ion configuration. The use of the long nanosecond pulse to generate the initial plasma column and establish the ionization conditions had the advantages of creating the x-ray amplifier pre-conditions using a small amount of laser energy. After a given delay, when the plasma density profile expands and relaxes to aid the propagation of the x-ray laser and secondly the absorption of the picosecond laser pulse, the main short pulse was fired. The short pulse rapidly heated the plasma raising the electron temperature and strongly pumping the upper level of the inversion by monopole electron excitation. The use of the two separate laser pulses to optimize the conditions for the x-ray laser was the main reason that the drive energy could be reduced. These were similar advantages to the pre-pulse technique that had been reported previously by Nilsen and colleagues [7].

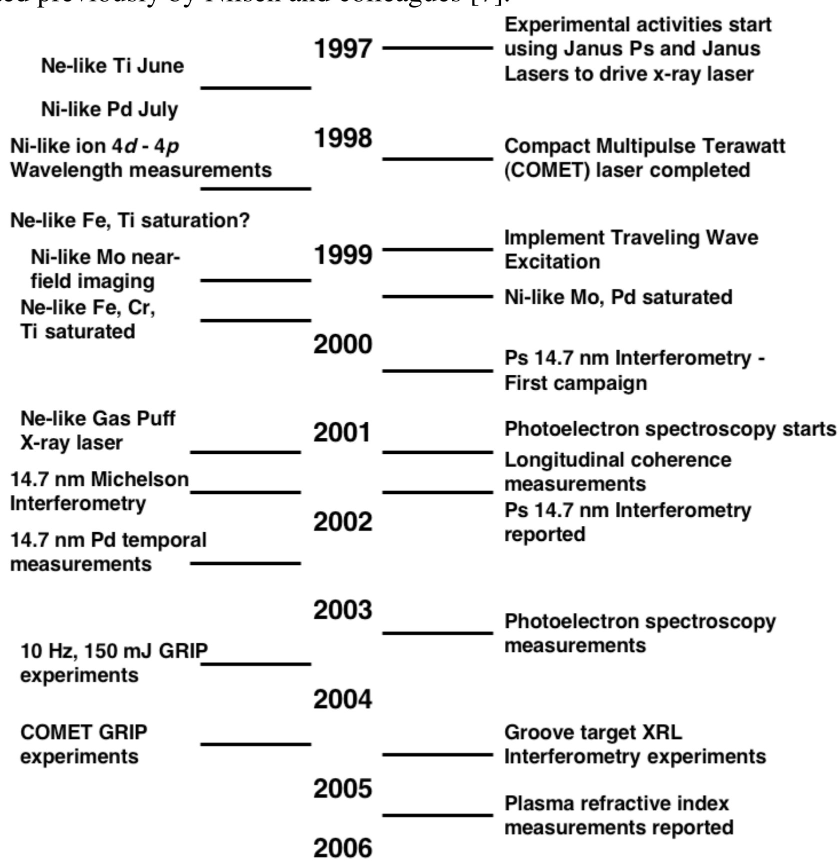

Fig. 1. Time-line showing major activities and achievements on the tabletop x-ray laser work at LLNL in the years 1997 through 2006.

The tabletop experimental activities first started at LLNL in January 1997 when the short pulse, tabletop Janus Picosecond Laser was available. There were several stages involved before the x-ray laser experiments could begin. The laser at the time was a single high power short pulse beam producing ~3 J at 1054 nm wavelength in a 500 fs pulse with a repetition rate of 1 shot/4 minutes. The beam diameter, along with the optimization of other parameters, was increased to improve the available compressed energy to 7.5 J giving the laser a 15 TW peak power specification. Initial experiments were conducted with a ~1.5 ps short pulse. The long pulse came from the Janus laser: Rod shots of 5 J energy at 1064 nm in a 800 ps (FWHM) pulse could be fired at 1 shot/3 minutes. The two lasers were synchronized to within 80 ps rms jitter relative timing. The beams had orthogonal polarization and were combined into one beam path with a polarizer, the long pulse in transmission and the short pulse in reflection [8, 9]. The single beam line to the target chamber was focused with a 4-m focal length plano-concave cylindrical lens and a 0.61-m focal length on-axis parabola. The laser line focus was 1.2 cm × 80 μm (L × W) and the two laser beams were imaged and carefully overlapped at the target plane.

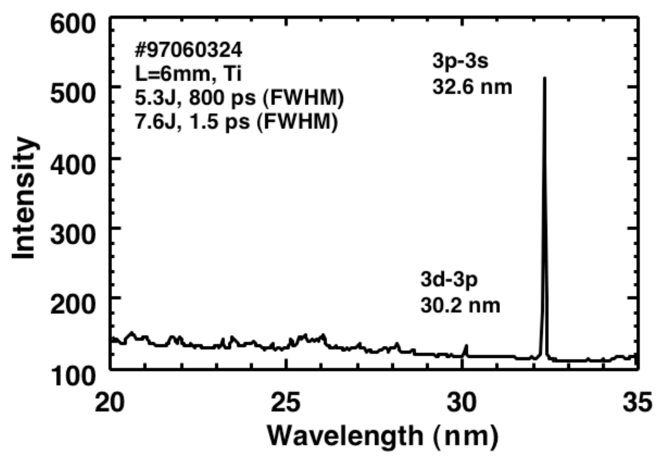

Fig. 2. First Ne-like ion x-ray laser spectrum observed for a 6 mm Ti target on 3 June 1997 using the Janus and Janus-ps lasers at LLNL [9].

The main diagnostic for the x-ray laser lines looking on-axis was a 1200 line mm^{-1} flat-field grating spectrometer with a back-thinned 1024 × 1024 charge-coupled device (CCD) detector to cover 13 - 35 nm. A gold-coated mirror collection optic imaged the plasma gain region with 1:1 magnification onto a 100 μm wide entrance slit. The instrument spatially resolved the position of the gain region. Additional instruments included a CCD x-ray slit camera with 25 μm spatial resolution for line focus uniformity and a CCD flat crystal KAP (001), 2d = 26.58 Å, spectrometer. All x-ray instruments used CCD detector systems that were LLNL designed, fabricated and optically calibrated.

The first targets used to study the Ne-like ion scheme were polished low- to mid-Z metals including Ti, Fe, and Ge. There was additional theoretical support for lasing on Ti under a range of different picosecond pulse durations [10]. The Ge target did not demonstrate lasing with the laser energies available. Initially Ti did not lase when the two laser pulses were separated by a delay of 800 ps peak-to-peak. Unexpectedly, some jitter in the laser synchronization delayed the short pulse to 1 ns which was sufficient to generate better plasma conditions for lasing [9]. Those first results are shown in Fig. 2 and further optimization of the x-ray signal was achieved with delays of 1.6 ns. The strongest lasing was on the 3*p* – 3*s* line at 32.6 nm with the 3*d* – 3*p* line also visible at ~5% intensity. Small signal gains of 24 cm^{-1} and gain length gL products approaching 15 for 1 cm targets were achieved [8]. Strong lasing was also observed on the Ne-like Fe 3*p* – 3*s* line at 25.5 nm. The main leap forward though was the demonstration of shorter wavelength lasing utilizing the Ni-like ion 4*d* – 4*p* scheme, specifically for Pd at 14.7 nm. The Ni-like schemes, with atomic number in the mid-forties, had not lased well under various pulse configurations on the larger lasers. There was a good match with the ps-laser pumping conditions here and gains of 35 cm^{-1} and gL products of 12.5 were reported [11]. One major difference between the Ne-like and Ni-like lasing was the lower output on the Ni-like schemes. This would be attributed to the shorter gain lifetime conditions for the Ni-like ion and would be improved by using traveling wave excitation.

3 Compact Multipulse Terawatt Laser

Early in 1998, the tabletop laser was redesigned to accommodate an additional 50 mm rod amplifier to generate a long stretched pulse beam with 600 ps (FWHM) at the full aperture of 8.4 cm diameter. This allowed the tabletop laser to generate two full energy ~7 J beams for the x-ray laser on two standard laser tables. The laser was renamed the Compact Multipulse Terawatt

(COMET) laser and was a stand-alone facility optimized for x-ray laser studies [12]. The vacuum compressor was upgraded with new 13" diameter, 1700 line mm^{-1}, gold-coated gratings. A series of experiments were conducted over the next few years to both characterize and improve the x-ray laser parameters required for applications. The careful measurement of various Ni-like $4d$ – $4p$ transition wavelengths was carried out in lasing plasmas and compared with optimized level multiconfiguration Dirac-Fock code as well as high resolution spectra from non-lasing vacuum spark or laser-produced plasmas [13]. This was largely to gain better understanding of the Ni-like ion energy level structure (that is complex) as well as the precise transition wavelength knowledge required to develop and optimize the reflectivity of x-ray optics. The Ni-like x-ray lasers also seemed to be a good match with the small ps-lasers and lower Z materials were observed to lase for the first time [14].

The next step was improving the output of a number of the Ne-like and Ni-like x-ray lasers. The Ne-like ion x-ray lasers studied to date were very close to saturation. A traveling wave excitation scheme was implemented before the final focusing optics utilizing a segmented reflection echelon that split the beam into delayed vertical slices [15]. This produced a segmented but contiguous line focus for both beams, initially with 5 steps then increased to 7 when the focus was lengthened to 1.6 cm. The traveling wave focus consisted of 0.22 cm long steps delayed by 7.7 ps relative to its neighbor. In addition the long pulse was defocused to 150-μm (FWHM) while retaining an 80-μm (FWHM) short pulse. This improved the x-ray laser propagation by reducing the negative effect of lateral density gradients. The combination of these two methods was very effective and increased the Pd x-ray laser output up to 100 times and into saturation for targets approaching 1 cm in length. Estimated output energy was now above 10 μJ and in saturation. Further optimization of the laser energy parameters and relative pulse delay gave narrow beam divergence ~2.5 mrad (FWHM) and improved x-ray brightness for the 14.7 nm laser. Ni-like Mo laser at 18.9 nm was driven into saturation [15] and multi-layer-coated imaging optics allowed direct measurements of the near-field and far-field patterns of both the Mo and Pd lasers [16, 17]. The speckle pattern and various structures observed in the far-field x-ray beam were recently attributed to a combination of low spatial coherence, high longitudinal coherence and short pulse duration of these transient x-ray sources [18].

One of the projects in 2001 was to study gas puff targets heated by transient pumping. The argon laser successfully lased on both $3p$ – $3s$ and $3d$ – $3p$ lines at 46.9 nm and 45.1 nm, respectively [19]. However, further work in studying the ionization and absorption conditions during both laser pulses would be useful to optimize the amplification process.

4 Applications, New source Development and Characterization

The Pd laser and the experimental setup described above became the work-horse for many of the studies up until the grazing incidence pumping (GRIP) method was adopted for the high repetition rate x-ray source development. In many cases the source characterization was in parallel with the application development. The plan for developing the x-ray laser interferometry of laser-produced plasmas based on the LLNL ps duration 14.7 nm line was discussed with Jorge Rocca's group at Colorado State University and a second French team led by Philippe Zeitoun in 1999. In the former case, a skewed Mach-Zehnder interferometer using diffraction grating optics as beam splitters was developed from the design used at 46.9 nm [20]. The instrument was robust, had a high signal throughput and achieved uniform fringes with high visibility. This Diffraction Grating Interferometer (DGI) was an excellent match for the tabletop transient x-ray lasers and led to new picosecond resolution and micron spatial resolution data from laser-produced plasma phenomena [21 – 23].

The use of the interferometry on Pd plasmas when combined with the near-field imaging technique of the gain region yielded quantitative measurements of the density profile of the x-ray laser amplifier medium [24]. Fig. 3(a) and (b) show the effect of different short pulse durations on x-ray amplification for the same pre-pulse conditions used to generate the 14.7 nm Pd laser [15]. This study showed the beneficial effect on the x-ray laser output of using longer 6 – 13 ps short pulse drives together with optimized laser parameters [24]. The experiment used various, sophisticated x-ray laser diagnostics to understand and achieve better amplification conditions.

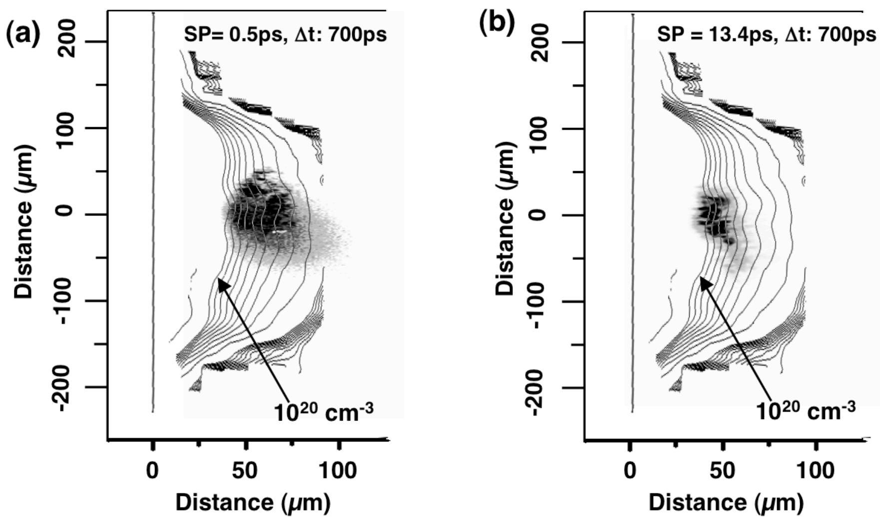

Fig. 3. The nearfield image of the Pd 14.7 nm x-ray laser, with both laser pulses fired, overlaid on a 2-D density map measured by 14.7 nm interferometry of the 600 ps heated Pd plasma at Δt = 700 ps. The short pulse would fire at this time. (a) Conditions for a 0.5 ps short pulse. (b) For a 13.4 ps short pulse where the x-ray laser output is substantially higher, more compact and less affected by refraction.

The second interferometry effort was based on a Michelson design using thin foil multilayer beamsplitters: A measurement of the longitudinal coherence of the Pd line operating in saturation was made for two different short pulse durations of ~6 ps, shown in Fig. 4(a), and ~13 ps [25]. This indicated that the Pd line was spectrally narrow approaching $\lambda/\Delta\lambda$ of 5×10^4. This observation is consistent with the high gain determined from various transient pumping experiments over the last decade. Another experiment was performed to measure the x-ray laser pulse duration of the Pd line under various laser parameters [26]. Figure 4 (b) shows the duration of the 14.7 nm under similar pumping conditions of Fig. 4(a). The measured duration is typically 4 – 5 ps when heated by a 6 ps pulse. Shorter pulse durations are achieved below 3 ps when the optical laser pulse duration is reduced or the x-ray laser is operated out of saturation. This is in close agreement with results reported for the Ag x-ray laser [27]. The spectral and temporal measurements allow us to note that the ps-driven Pd laser is operating in the few times transform-limited regime [28] in agreement with similar work as reported by other groups [29 – 31].

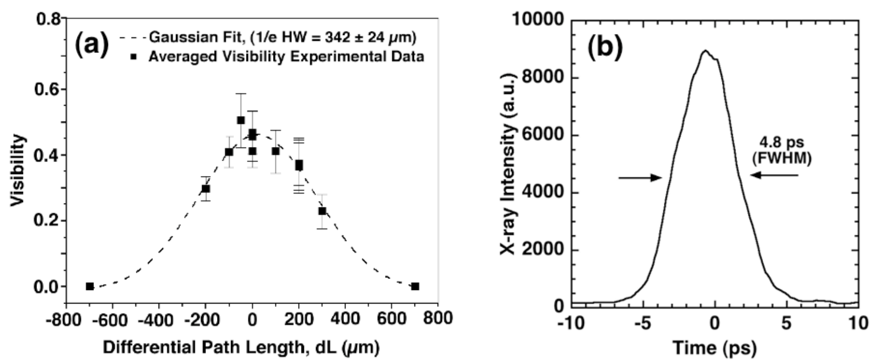

Fig. 4. (a) Fringe visibility of saturated Pd x-ray laser line heated by a 6 ps short pulse measured as a function of the differential arm separation of the Michelson interferometer. (b) Pulse duration of Pd 14.7 nm laser line measured with a fast 500 fs x-ray streak camera under similar heating and saturated output conditions.

We mention some recent activities. The Pd laser was shown to be a novel materials surface probe where the x-ray laser, incident on the surface, can photo-ionize the electrons in low energy valence and shallow core levels. The resultant kinetic energy of the ejected electrons is measured using time-of-flight spectroscopy to retrieve the original electron energy level structure [32]. This allows a ps snapshot of the electronic structure of the material while undergoing rapid changes [32]. A stable and high repetition soft x-ray laser source would have advantages for this type of work since large photon numbers in single bursts are not necessary. High repetition rate ~10 Hz lasing has been demonstrated in our laboratory using the grazing incidence pumping geometry where the required laser pump energy can be reduced to the range of 150 mJ to ~1 J [33]. This scheme has been quickly adopted in numerous laboratories and is presented in various papers in this journal. A photoelectron microscopy tool based on high repetition, fs-heated vacuum ultraviolet radiation has been utilized for detailed surface chemistry studies [34]. This is one area where the higher photon energy and more monochromatic energy of the x-ray laser would make a strong application.

5 Conclusions

We have given a brief overview of the last 10 years of research in tabletop x-ray lasers at LLNL. With the development of the Compact Multipulse Terawatt (COMET) laser as a dedicated x-ray laser facility a number of advances were made in the source development, including optimization of the output, optimization of key parameters and establishment of several applications. In this time several studies were conducted that broke new ground for ps-driven x-ray lasers: The gas puff x-ray laser at 46.9 nm illustrated the potential for a debris-less gain medium. The Grazing Incidence Pumping (GRIP) was shown to operate with laser energy as low as 150 mJ and is the route forward for the development of a high average power, high repetition rate 10 – 30 nm source. The collisional excitation scheme has been the continuous link through this research and will likely continue to in the near future.

Acknowledgments

Work performed under the auspices of the US Department of Energy by the University of California Lawrence Livermore National Laboratory under Contract No. W-7405-Eng-48 and in part by US Department of Energy grant No. #DE-FG52-06NA26152, and in part, by the Ministry of Science and High Education of Poland under grant No 3 T11B 031 26 and by the NATO Collaborative Grant PST.CLG.979339.

References

1. J. J. Rocca *et al*., *Phys. Rev. Lett.* **73**(16), 2192 - 2195 (1994).
2. J. J. Rocca *et al*., *Phys. Rev. Lett.* **77**(8), 1476 - 1479 (1996).
3. B. E. Lemoff *et al*., *Phys. Rev. Lett.* **74**(9), 1574 - 1577 (1995).
4. P. V. Nickles *et al*., *SPIE Proceedings* **2520**, 373 - 378 (1995); P. V. Nickles *et al*., *Phys. Rev. Lett.* **78**(14), 2748 - 2751 (1997).
5. D. V. Korobkin *et al*., *Phys. Rev. Lett.* **77**(26), 5206 - 5209 (1996).
6. V. N. Shlyaptsev *et al*., *SPIE Proceedings* **2012**, 111 - 118 (1993).
7. J. Nilsen, B. J. MacGowan, L. B. Da Silva, and J. C. Moreno, *Phys. Rev. A* **48**(6), 4682 - 4685 (1993).

8. J. Dunn, A. L. Osterheld, R. Shepherd, W. E. White, V. N. Shlyaptsev, A. B. Bullock, and R. E. Stewart, *SPIE Proceedings* **3156**, 114 - 121 (1997).
9. J. Dunn, A. L. Osterheld, R. Shepherd, W. E. White, V. N. Shlyaptsev, and R. E. Stewart, "Table-Top Transient Collisional Excitation X-ray Laser Research at LLNL: Status June 1997", Lawrence Livermore National Laboratory, Livermore, CA UCRL-ID-127872 (1997).
10. J. Nilsen, *Phys. Rev. A* **55**(4), 3271 - 3274 (1997).
11. J. Dunn, A. L. Osterheld, R. Shepherd, W. E. White, V. N. Shlyaptsev, and R.E. Stewart, *Phys. Rev. Lett.* **80**, 2825 - 2828 (1998).
12. COMET has 4 independent, synchronized beams: The two main beams, a third ~20 mJ (1ω or 2ω) short pulse probe beam and a fourth beam that uses energy from the long pulse arm for either 3 J in 600 ps or ~2 J compressed.
13. Y. Li, J. Nilsen, J. Dunn, A. L. Osterheld, A. Ryabtsev, and S. Churilov, *Phys. Rev. A* **58**, R2668 - 2671 (1998).
14. J. Dunn *et al.*, *Opt. Lett.* **24**, 101 – 3 (1999).
15. J. Dunn, Y. Li, A. L. Osterheld, J. Nilsen, J. R. Hunter, V. N. Shlyaptsev, *Phys. Rev. Lett.* **84**, 4834 - 7 (2000).
16. Y. L. Li, J. Dunn, J. Nilsen, T. W. Barbee, Jr., A. L. Osterheld, and V. N. Shlyaptsev, *J. Opt. Soc. Am. B* **17**, 1098 - 1101 (2000).
17. J. Nilsen *et al.*, *J. Opt. Soc. Am. B* **20**, 191 - 194 (2003).
18. O. Guilbaud, A. Klisnick, K. Cassou, S. Kazamias, D. Ros, G. Jamelot, D. Joyeux, and D. Phalippou, *Europhys. Lett.* **74**, 823 – 829 (2006).
19. H. Fiedorowicz, A. Bartnick, J. Dunn, R. F. Smith, J. R. Hunter, J. Nilsen, A. L. Osterheld, and V. N. Shlyaptsev, *Opt. Lett.* **26**, 1403 - 1405 (2001).
20. J. Filevich, K. Kanizay, M. C. Marconi, J. L. A. Chilla, and J. J. Rocca, *Opt. Lett.* 25, 356 - 358 (2000).
21. R. F. Smith *et al.*, *Phys. Rev. Lett.* **89**(6), 065004-1 (2002).
22. J. Filevich *et al.*, *Appl. Opt.* **43**(19), 3938 - 46 (2004).
23. J. Filevich *et al.*, *Phys. Rev. Lett.* **94**, 035005 (2005).
24. R. F. Smith *et al.*, *Phys. Rev. E* **72**, 036404 (2005).
25. R. F. Smith *et al.*, *Opt. Lett.* **28**, 2261 - 3 (2003).
26. J. Dunn *et al.*, *SPIE Proceedings* **5197**, 51 - 59 (2003).
27. A. Klisnick *et al.*, *Phys. Rev. A.* **65**, 033810 (2002).
28. J. Dunn *et al.*, *SPIE Proceedings* **5197**, 43 - 50 (2003).
29. Y. Abou-Ali *et al.*, *Opt. Comm.* **215**, 397 - (2003).
30. A. Klisnick *et al.*, *J. Quant. Spectrosc. Radiat. Transf.* **99**, 370 – 380 (2006).
31. D. Benredjem et al., *Phys. Rev. A* **73**, 063820 (2006).
32. A. J. Nelson *et al.*, *Appl. Phys. Lett.* **87**, 154102 (2005).
33. R. Keenan *et al.*, *Phys. Rev. Lett.* **94**, 103901 (2005).

34. T. Munakata *et al*., *Surf. Sci,* **532**, 1140 – 144 (2003).